# Variational analyses of series expansions for the exchange-interaction model

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The linked-cluster type of series expansions with a variational parameter are derived to the eighth order for the spin-$S$ exchange-interaction model. These series expansions are analyzed to obtain phase transition temperatures $T_c$, latent heats, and discontinuities of the order parameter at $T_c$ for the face-centered-cubic lattice.

*I. Introduction.* The exchange operator studied by Schrödinger[1] in 1941 had been used to construct a spin model, called the exchange-interaction (EI) model,[2–5] which contains multipole interactions of the form $(S_i \cdot S_j)^n$ with $n=1,2,\ldots,2S$. This model attracts wide theoretical interests, and pedagogic attentions. The complexities which arise from the nonlinearity and the noncommutability of quantum spin operators prohibit the model from deep understanding of its thermodynamic properties.

When the spin $S=1/2$, the EI model reduces to the Heisenberg[6] model, which has been well studied. For spin-1 systems which contain dipole and quadrupole interactions, critical properties have been studied by various methods.[7–9] For general spins ($S>1$), limited results are obtained from the studies of high-temperature series expansions,[10] real-space renormalizations,[11,12] quantum Monte Carlo simulations,[13] and the Green function method.[14] All of these studies for $S>1$ did not investigate thermal variations of the order parameters. Besides, the system was assumed to have a continuous phase transition. Recently great efforts have been made in effective-field theories, such as the mean-field approximation,[15,16] the finite cluster approximation,[17] and the constant coupling approximation.[18] All of the effective-field studies show that the EI model undergoes a first-order phase transition for $S \geqslant 1$.

It has been shown[15] that in the mean-field approximation, the EI model has exactly the same critical properties as the Potts model. Both models undergo a first-order phase transition for $S \geqslant 1$. Determinations of the critical parameters for first-order phase transitions have been more difficult and less successful than those for continuous transitions. We have recently developed a method[19] of analyzing critical properties from the linked-cluster series expansions. Very good results have been obtained when the method is applied to the Potts model which is known to have a first-order phase transition. The mean-field theory is the lowest-order approximation of our method.

In this Brief Report we study the critical properties of the EI model for general spins by using the same method proposed in Ref. 19. We derive the free energy series expansions similar to the linked-cluster series expansions, but treat the parameter involved as a variational one. In Sec. II we describe briefly the method to calculate the free energy series expansions, and derive formulas for the order parameter and the internal energy. In Sec. III series expansions to the eighth order are analyzed to obtain the critical temperatures $T_c$, the discontinuities of the order parameters at $T_c$, denoted as $\Delta M$, and the latent heat, denoted as $\Delta U$ for the face-centered-cubic lattice. A brief discussion is also made in Sec. III. Finally, some of the important calculations for the semi-invariants are given in the Appendix.

*II. Linked-cluster series expansions for the EI model.* The Hamiltonian of the EI model can be expressed[16] as

$$-\beta H=(J/kT)\sum_{\langle ij\rangle} P_{ij} =K\sum_{\langle ij\rangle}\sum_{l=0}^{2S}\sum_{m=-l}^{l} A(S,l)Q_m^{(l)}(S_i)Q_m^{(l)}(S_j), \quad (1)$$

where $\beta=1/kT$, $J$ is the coupling constant, $K=\beta J$, and the summation $\Sigma_{\langle ij\rangle}$ is over all nearest-neighbor pairs of spins. The exchange operator $P_{ij}$ is to permute the spin variables $S_i$ and $S_j$:

$$P_{ij}f(S_i,S_j)=f(S_j,S_i)P_{ij}, \quad (2)$$

for any spin-state function $f(S_i,S_j)$. The coefficients $A(S,l)$ are[15]

$$A(S,l)=2^{2l}(2l+1)(2S-l)!/(2S+l+1)!, \quad (3)$$

and $Q_m^{(l)}$ are spin multipole moments given as

$$Q_0^{(0)}=1, \quad Q_0^{(1)}=S_z, \quad Q_1^{(1)}=S_x, \quad Q_{-1}^{(1)}=S_y,$$

$$Q_0^{(2)}=(3/2)[S_z^2-S(S+1)/3], \quad Q_1^{(2)}=\sqrt{3}/2(S_xS_z+S_zS_x),$$

$$Q_{-1}^{(2)}=\sqrt{3}/2(S_yS_z+S_zS_y), \quad Q_2^{(2)}=\sqrt{3}/2(S_x^2-S_y^2),$$

$$Q_{-2}^{(2)}=\sqrt{3}/2(S_xS_y+S_yS_x), \text{ etc.} \quad (4)$$

All of the spin multipole moments $Q_m^{(l)}$ for $l\neq 0$ are traceless and Hermitian operators.

For a system of $N$ spins on a lattice of coordination number $z$, the mean-field Hamiltonian[16] $H_M$ is

$$-\beta H_M=Kz\sum_{i=1}^{N}\sum_{l=0}^{2S}\sum_{m=-l}^{l} A(S,l)Q_m^{(l)}(\mathbf{S}_i)\langle Q_m^{(l)}\rangle - \frac{NKz}{2}\sum_{l=0}^{2S}\sum_{m=-l}^{l} A(S,l)\langle Q_m^{(l)}\rangle^2, \quad (5)$$

where $\langle Q_m^{(l)}\rangle$ is the mean-field thermal average of $Q_m^{(l)}$. The order parameter $M(T)$, also called polarization, is defined[16] as

 

$$\langle Q_m^{(l)}\rangle = M(T)\langle\phi_k|Q_m^{(l)}|\phi_k\rangle, \quad (l\neq 0), \tag{6}$$

where $|\phi_k\rangle$ is the ground state of the system. The polarization $M(T)$ has been shown[9,20] to be independent of $l,m$ and $|\phi_k\rangle$. All multipole moments $Q_m^{(l)}(l\neq 0)$ are equivalent, and $M(T)$ is the only order parameter of the system.

The second term in Eq. (5) is a constant operator which can be neglected. The mean-field Hamiltonian reduces to

$$-\beta H_M = KzM\sum_{i=1}^{N}\rho_k(\mathbf{S}_i), \tag{7}$$

with the spin density matrix

$$\rho_k(\mathbf{S}_i)=\sum_{l=0}^{2S}\sum_{m=-1}^{l}A(S,l)\langle\phi_k|Q_m^{(l)}|\phi_k\rangle Q_m^{(l)}(\mathbf{S}_i). \tag{8}$$

As shown in Ref. 16, for any set of $2S+1$ orthonormal single-spin states: $|\phi_0\rangle,|\phi_1\rangle,\ldots,|\phi_{2S}\rangle$, $\rho_k=|\phi_k\rangle\langle\phi_k|$, and

$$\rho_k|\phi_j\rangle = |\phi_k\rangle\langle\phi_k|\phi_j\rangle = \delta_{kj}|\phi_j\rangle. \tag{9}$$

Therefore $\rho_k$ has only one nonzero matrix element: $\langle\phi_k|\rho_k|\phi_k\rangle=1$. All other matrix elements vanish. This property simplifies greatly the trace calculations when $\rho_k$ are involved.

In our derivation of the linked-cluster series expansions, we define a single-spin Hamiltonian $H_0$ which is obtained by replacing $KzM$ in the mean-field Hamiltonian by a variational parameter $L$, i.e.,

$$-\beta H_0 = L\sum_{i=1}^{N}\rho_k(\mathbf{S}_i). \tag{10}$$

The Hamiltonian is rewritten as

$$H = H_0 + H_1, \tag{11}$$

with the fluctuation Hamiltonian $H_1$ given by

$$-\beta H_1 = K\sum_{\langle ij\rangle}P_{ij} - L\sum_{i=1}^{N}\rho_k(\mathbf{S}_i). \tag{12}$$

The partition function is then written as

$$Z = \mathrm{Tr}\,\exp(-\beta H) = Z_0\langle\exp(-\beta H_1)\rangle_0, \tag{13}$$

where $\langle\ \rangle_0$ is the thermal average with respect to the Hamiltonian $H_0$ and

$$Z_0 = \mathrm{Tr}\,\exp(-\beta H_0) = (e^L+2S)^N. \tag{14}$$

The mean-field free energy per spin is

$$-\beta F_0/N = N^{-1}\ln Z_0 = \ln(e^L+2S). \tag{15}$$

The linked-cluster series expansions of the correction to the mean-field free energy $\Delta F\equiv F-F_0$ is

$$-\beta\Delta F(K,L)/N = N^{-1}\ln(Z/Z_0) = N^{-1}\ln\langle\exp(-\beta H_1)\rangle_0 = \frac{1}{N}\ln\left\{1+\sum_{n=1}^{\infty}\left\langle\left[K\sum_{\langle ij\rangle}P_{ij}-L\sum_{i}^{N}\rho_k(\mathbf{S}_i)\right]^n\right\rangle_0 \Big/ n!\right\}$$

$$= \frac{1}{N}\sum_{n=1}^{\infty}\left\langle\left[K\sum_{\langle ij\rangle}P_{ij}-L\sum_{i}^{N}\rho_k(\mathbf{S}_i)\right]^n\right\rangle_c \Big/ n! = \sum_{n=1}^{\infty}\sum_{m=0}^{n}a_{nm}K^{n-m}L^m/n!. \tag{16}$$

Here $\langle\ \rangle_0$ is the thermal average, while $\langle\ \rangle_c$ is the cumulant average. The coefficients $a_{nm}$ can be calculated by the diagrammatic method, or by the cluster expansion method.[6,21] The main difference between the present calculation and the previous work for the Potts model[19] is that Kronecker $\delta$ functions $\delta_{ij}$ in the Potts model are replaced by spin exchange operators $P_{ij}$. Since $P_{ij}$ and $P_{jk}$ do not commute, the present calculation is much more complicated than that of the Potts model. Some of the important thermal averages which involve products of $P_{ij}$ and $\rho_k$ are given in the Appendix.

If the summation is taken up to the $n$th order, we obtain the $n$th-order free energy, denoted as $F^{(n)}$. The first-order and the second-order free energies depend only on the coordination number $z$. They are

$$-\beta F^{(1)}/N = N^{-1}\ln Z_0 + (zD_2/2)K - xL, \tag{17}$$

$$-\beta F^{(2)}/N = -\beta F^{(1)}/N + [z^2(D_3-D_2^2) + z(D_2^2-2D_3+1)/2]K^2/2 + zx(D_2-x)KL + (x-x^2)L^2/2, \tag{18}$$

with $x\equiv e^L/(e^L+2S)$ and $D_n\equiv(e^{nL}+2S)/(c^L+2S)^n$. $F^{(3)}$ and higher-order free energies depend on the details of the lattice structure. We have calculated the coefficients $a_{nm}$ to the eighth order ($m\leqslant n\leqslant 8$) numerically for the cubic lattices. It is too lengthy to present these coefficients in this article. If $L=0$, Eq. (16) reduces to the high-temperature series expansion; and if $L=Kz[\exp(KzM)-1]/[\exp(KzM)+2S]$, Eq. (16) is the same as the conventional linked-cluster series expansion. In the present method, $L$ is treated as a variational parameter, and the stable value of $L$ is determined by minimizing the free energy.

The parameter $L$ is related to the thermal average $\langle Q_m^{(l)}\rangle$, and is nonzero if and only if the system is ordered.

Therefore, the stable value of $L$ may be considered as the order parameter of the system. However, it is important to note that $L$ is not the same as the conventional order parameter $M$ defined in Eq. (6), which is proportional to the thermal averages of the spin multipole moments. The order parameter $M$ can be derived from the thermal average of the spin density operator. We have

$$\langle \rho_k \rangle = \sum_{l=0}^{2S} \sum_{m=-l}^{l} A(S,l) \langle \phi_k | Q_m^{(l)} | \phi_k \rangle \langle Q_m^{(l)} \rangle$$

$$= M \sum_{l=1}^{2S} \sum_{m=-l}^{l} A(S,l) \langle \phi_k | Q_m^{(l)} | \phi_k \rangle^2 + (2S+1)^{-1}$$

$$= (2SM+1)/(2S+1). \tag{19}$$

Uses have been made of Eq. (6) and the relation[16]

$$\sum_{l=0}^{2S} \sum_{m=-l}^{l} A(S,l) \langle \phi_k | Q_m^{(l)} | \phi_k \rangle^2 = 1. \tag{20}$$

We also have

$$\left\langle \sum_i \rho_k(S_i) \right\rangle = \frac{\mathrm{Tr} \Sigma_i \rho_k(S_i) e^{-\beta H}}{\mathrm{Tr} e^{-\beta H}} = \frac{\langle \Sigma_i \rho(S_i) e^{-\beta H_1} \rangle_0}{\langle e^{-\beta H_1} \rangle_0}$$

$$= -\frac{\partial}{\partial L} \ln \langle e^{-\beta H_1} \rangle_0 . \tag{21}$$

Therefore

$$-\frac{2SM+1}{2S+1} = \frac{1}{N} \frac{\partial}{\partial L} \ln \langle e^{-\beta H_1} \rangle_0$$

$$= \sum_{n=1}^{\infty} \sum_{m=1}^{n} m a_{nm} K^{n-m} L^{m-1} / n! . \tag{22}$$

The coefficients $a_{nm}$ are functions of $L$ as illustrated in Eqs. (17) and (18). But in the above equation $Z_0$ and $a_{nm}$ are regraded as constants, when the derivative of $-\beta \Delta F$ with respect to $L$ is calculated.

Similarly the internal energy per spin of the system is given by

$$-\frac{U}{NJ} = \frac{\langle \Sigma_{\langle ij \rangle} P_{ij} \rangle}{N} = \frac{1}{N} \frac{\mathrm{Tr} \Sigma_{\langle ij \rangle} P_{ij} e^{-\beta H}}{\mathrm{Tr} e^{-\beta H}}$$

$$= \frac{1}{N} \frac{\langle \Sigma_{\langle ij \rangle} P_{ij} e^{-\beta H_1} \rangle_0}{\langle e^{-\beta H_1} \rangle_0} = \frac{1}{N} \frac{\partial}{\partial K} \ln \langle e^{-\beta H_1} \rangle_0$$

$$= \sum_{n=1}^{\infty} \sum_{m=0}^{n-1} (n-m) K^{n-m-1} L^m / n! . \tag{23}$$

For the $n$th-order free energy series, the corresponding order parameter $M^{(n)}$ and the internal energy $U^{(n)}$ are one order lowered.

*III. Results and discussions.* We consider $F^{(n)}(K,L)$ as the Landau free energy of the system, and $L$ as the Landau order parameter. One can plot $F^{(n)}$ versus $L$ at a given temperature. The value of $L$ which gives the lowest free energy $F^{(n)}$ is the stable value of $L$ at the given temperature, denoted as $L_s^{(n)} \times (K)$. We calculate $L_s^{(n)}$ as functions of the temperature numerically. At low temperatures, the free energy curves have only one minimum at $L_s^{(n)} > 0$. It indicates that the system is in the ordered phase. At high temperatures, minimum occurs at $L_s^{(n)} = 0$ only and the system is disordered. For temperatures in between, the free energy curves have one maximum which is unstable and two minima: one is zero and the other is positive. At the critical temperature $T_c^{(n)}$, both minima have the same free energy. Above $T_c^{(n)}$, the positive minimum is a metastable phase. The free energy analysis shows that the EI model undergoes a first-order phase transition for $S \geqslant 1$ as predicted by the effective-field approximations.[15–18] For spin-1/2 system, the bifurcation of the two minima does not occur. It undergoes a continuous phase transition. Results of $T_c^{(n)}$ are given in Table I.

TABLE I. Critical temperatures $kT_c^{(n)}/J$ for the face-centered-cubic lattice. HTS means the high temperature series expansion (Ref. 10) MFA represents the mean-field approximation (Ref. 16).

| $n$ | $S=1/2$ | $S=1$ | $S=3/2$ | $S=2$ | $S=5/2$ |
|---|---|---|---|---|---|
| MFA | 6 | 4.3248 | 3.6410 | 3.2461 | 2.9824 |
| 1 | 6 | 4.3248 | 3.6410 | 3.2461 | 2.9824 |
| 2 | 5 | 3.5836 | 2.9873 | 2.6416 | 2.4100 |
| 3 | 23/5 | 3.3880 | 2.8634 | 2.5555 | 2.3479 |
| 4 | 4.4293 | 3.3468 | 2.8580 | 2.5655 | 2.3662 |
| 5 | 4.3494 | 3.3160 | 2.8275 | 2.5300 | 2.3252 |
| 6 | 4.3003 | 3.2812 | 2.7926 | 2.4940 | 2.2879 |
| 7 | 4.2621 | 3.2605 | 2.7840 | 2.4953 | 2.2981 |
| 8 | 4.2306 | 3.2521 | 2.7822 | 2.4967 | 2.3018 |
| $\infty$ | 4.02 | 3.139 | 2.708 | 2.442 | 2.250 |
| HTS | 4.02 | 3.10 | 2.64 | 2.35 | 2.14 |

It is not clear theoretically how the $n$th-order critical temperatures converge to the limiting value. Similar to the Potts model[19] we assume that

$$T_c^{(n)} = T_c + c n^{-1} + O(n^{-2}). \tag{24}$$

Extrapolation of the $n$th-order critical temperatures to $n \to \infty$ yields the phase transition temperature $T_c$ of the system. Our results based on the series expansions up to the eighth order are shown in Table I. The uncertainties in our estimates of $T_c$ are within one percent for small spins. The values estimated by the high-temperature series expansion[10] are also included for comparison.

Using Eqs. (22) and (23), we calculate the $n$th-order approximations of the order parameter $M^{(n)}$ and the internal energy $U^{(n)}$, evaluated at $L = L_s^{(n)}(K)$ for the given temperature. The discontinuities of $M^{(n)}$ and $U^{(n)}$, denoted as $\Delta M^{(n)}$ and $\Delta U^{(n)}$ which are evaluated at $T_c^{(n)}$ are listed in Tables II and III, respectively. Similar extrapolations as Eq. (24) are also made to obtain the limiting values of the critical parameters. The uncertainties in our estimates of $\Delta M$ and $\Delta U$ are within a few percent.

The series of $M^{(n)}$ and $U^{(n)}$ are one order shorter than the free energy series. Besides, $\Delta M^{(n)}$ and $\Delta U^{(n)}$ fluctuate with $n$ vigorously for large spins. The accuracies in the extrapolation of the limiting values become poor when $S > 5/2$.

For the face-centered-cubic lattice, the free energy series up to the eighth order behaves so smooth that the critical

TABLE II. Discontinuities of the order parameters $\Delta M^{(n)}$ for the face-centered-cubic lattice.

| $n$ | $S=1$ | $S=3/2$ | $S=2$ | $S=5/2$ |
|---|---|---|---|---|
| MFA | 1/2 | 2/3 | 3/4 | 4/5 |
| 1 | 1/2 | 2/3 | 3/4 | 4/5 |
| 2 | 0.4889 | 0.6592 | 0.7509 | 0.8112 |
| 3 | 0.4294 | 0.5827 | 0.6690 | 0.7281 |
| 4 | 0.3644 | 0.4990 | 0.5751 | 0.6260 |
| 5 | 0.3436 | 0.4959 | 0.5905 | 0.6609 |
| 6 | 0.3500 | 0.5128 | 0.6190 | 0.7054 |
| 7 | 0.3435 | 0.4929 | 0.5841 | 0.6510 |
| 8 | 0.3144 | 0.4428 | 0.5139 | 0.5563 |
| ∞ | 0.273 | 0.387 | 0.486 | 0.55 |

TABLE III. Latent heat per spin $\Delta U^{(n)}/NJ$ for the face-centered-cubic lattice.

| $n$ | $S=1$ | $S=3/2$ | $S=2$ | $S=5/2$ |
|---|---|---|---|---|
| MFA | 1 | 2 | 27/10 | 16/5 |
| 1 | 1 | 2 | 27/10 | 16/5 |
| 2 | 0.6565 | 1.3216 | 1.8022 | 2.1620 |
| 3 | 0.4328 | 0.9319 | 1.3393 | 1.6750 |
| 4 | 0.3025 | 0.7033 | 1.0552 | 1.3540 |
| 5 | 0.2591 | 0.6619 | 1.0350 | 1.3643 |
| 6 | 0.2523 | 0.6545 | 1.0343 | 1.3860 |
| 7 | 0.2334 | 0.5996 | 0.9461 | 1.2625 |
| 8 | 0.1942 | 0.4988 | 0.7804 | 1.0198 |
| ∞ | 0.037 | 0.273 | 0.534 | 0.81 |

temperatures $T_c^{(n)}$ converge rapidly. For other cubic lattices whose lattice structure are not so compact as the face-centered-cubic lattice, the free energy series up to the eighth order do not give smooth $T_c^{(n)}$. Analyzing such series for critical parameters become unreliable even for small spins.

In general, the successive orders of critical parameters converge more rapidly for small spins. This manifest that for high spins the nonlinearities $(\mathbf{S}_i\cdot\mathbf{S}_j)^n$ interact in complicated ways. In conclusion our studies for the face-centered-cubic lattice confirm that the EI model undergoes a first-order phase transition for $S\geqslant 1$.

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*Appendix.* In calculating the series expansions, we have to evaluate

$$\langle A\rangle_0=\frac{\mathrm{Tr}\ A\ \exp(-\beta H_0)}{\mathrm{Tr}\ \exp(-\beta H_0)}, \tag{A1}$$

where $A$ are products of exchange operators $P_{ij}$ and density matrices $\rho_k(\mathbf{S})$. Since the products of $P_{ij}$ can be expressed in terms of the product of independent permutation cycles $(i_1, i_2,...,i_p)$, and each cycle can be evaluated independently in the trace calculation, all calculations of $\langle A\rangle_0$ can be decomposed into products of $\langle(i_1,i_2,...,i_p)\rho_k^{n_{i_1}}(\mathbf{S}_{i_1})\rho_k^{n_{i_{22}}}\times(\mathbf{S}_{i_2})\cdots\rho_k^{n_{i_p}}(\mathbf{S}_{i_p})\rangle_0$, $\langle(i_1,i_2,...,i_p)\rangle_0$, and $\langle\rho_k^{n_i}(\mathbf{S}_i)\rangle_0$. Here at least one of the integers $\{n_i\}_{i=i_1}^{i_p}$ is nonzero. By using the permutation property of $P_{ij}$ and the property[15] of the density matrices $\rho_k$, one can show that

$$\langle(i_1,i_2,\ldots,i_p)\rho_k^{n_{i1}}(\mathbf{S}_{i_1})\rho_k^{n_{i_2}}(\mathbf{S}_{i_2})\cdots\rho_k^{n_{i_p}}(\mathbf{S}_{i_\rho})\rangle_0=x^p, \tag{A2}$$

$$\langle i_1,i_2,\ldots,i_p\rangle_0=(e^{pL}+2S)/(e^L+2S)^p\equiv D_p, \tag{A3}$$

$$\langle\rho_k^{n_i}(\mathbf{S}_i)\rangle_0=x,\quad (n_i\neq 0), \tag{A4}$$

with $x\equiv e^L/(e^L+2S)$.

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